acp-2020-1086 (Walden et al.)
Measurement report: Characterization of uncertainties of fluxes and fuel sulfur content from ship emissions at the Baltic Sea

Reply to Editor

We thank the Editor for the positive feedback. We have made a minor revision and replied to all the comments. Our answers below are written by blue color. Additionally, language was checked and some clarifications were added. The changes made in the manuscript are written below by red color, and highlighted in the manuscript by yellow color.

Technical Comments:
page 5, line 135: Marine EC CO2 measurements are difficult. The methods are important, and measured fluxes can be off by orders of magnitude and of the wrong sign if methods are not optimal. Most EC measurements at sea that do not actively remove water vapor fluctuations from the air stream have been discredited. This manuscript does not provide evidence that their EC CO2 flux measurements are sound. How large was the Webb correction term compared to the actual CO2 flux (they say it is small but did not say how small)?

We agree that marine EC CO2 measurements are complicated, see also the answer to Editor's comments (below) concerning page 10, line 295.
We added a new figure in the SI (Fig. S5) to show the WPL correction for water vapor with respect to uncorrected CO_2 flux in both campaigns at Harmaja 2011 and Harmaja 2012. Correction for temperature was omitted as described in the text (p. 15, line 440). As can be seen, during the period of 28.8 to 31.8 the WPL correction for water vapor is significant changing even the sign of the CO_2 flux. At Harmaja 2012 the influence of the WPL correction was clearly less than in the campaign in 2011. In the attached figure the period when the air masses were arriving from the Atlantic Ocean, i.e. 28.8. to 31.8., the latent heat flux increased 3 to 6 times compared to rest of the period and explained the change of the sign of the CO_2 flux.

The following test was added in:
page 15, lines 458-461. The WPL correction performed to both of the data in 2011 and 2012 was small in general, see Fig S5. In the 2011 the sign of the flux was changed after correction of WPL during the period between 28.8 and 31.8.

page 8, line 235: how did the EC CO2 fluxes from the LI7000 compare with the Picarro?

The EC CO_2 fluxes were measured only by LI7000. We used Picarro G2301 for gradient technique together with LI7000 since the model G2301 is not fast enough for EC technique. This was mentioned in Section 3.2, page. 8, lines 238-240.

page 10, line 295: R/V Aranda LI7200 EC CO2 measurements were made on an undried air stream, relying instead on the Webb correction. I'm not aware of credible EC CO2 flux measurements made using an undried LI7200. There are several papers on this topic now (eg, Landwehr et al 2014, and others).

In our paper Sahlée et al. (2011) we compared the WPL corrected open path to the closed path sensor mounted on the same mast on R/V Aranda. The results were good for the small fluxes, under 0.5x10-3 mmol/m3*m/s. In the present study the LI-7200 fluxes were under that limit. According to Landwehr et al 2014 the flux estimates between the open path and closed path instruments agree well when the latent heat is less than 7 W m^{-2}. During the campaign R/V Aranda was near Harmaja in 2012 the latent heat flux was lower than this limit most of the time, see attached figure.

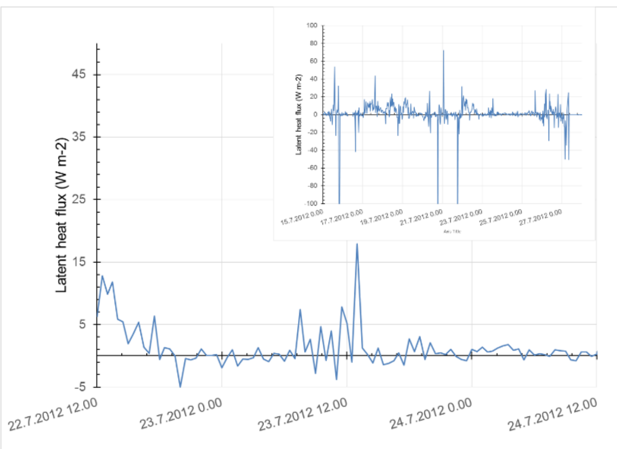

Figure. Latent heat flux during Harmaja 2012 campaign. The small figure presents the latent heat flux during the whole measurement campaign and the bigger figure during the joint measurements by R/V Aranda.

As pointed out by the reviewer (comment on Fig 12b), the pCO_2 difference was large during the R/V Aranda measurements, well over the flux detection limits suggested by Blomquist et al. (2014). Also the ship movements inside the archipelago were small and could not be detected from the LI-7200 timeseries (Miller et al. 2010).

However, there is a source of uncertainty in the R/V Aranda CO_2 flux measurements: the flow rate was low (10 slpm), and the fluxes might be underestimated. The fluxes in the beginning of the period agreed well with the open path LI-7500 at the same height on the mast which gives confidence on the LI-72000 measurements. After rain showers in the middle of the period, the LI-7500 showed much higher values that might be due the wet lens on the LI-7500. The LI-7200 is better shielded from the rain.

The magnitude of the R/V Aranda LI-7200 fluxes also compared rather well to the CO_2 fluxes measured at Harmaja where the measuring system was comparable to LI-7200, using humid air.

We added a sentence into:
page.15, lines 453-457: The corrections to CO_2 fluxes measured from humid air samples have been criticized to be insufficient (e.g. Landwehr et al. 2014). The fluxes in this study mainly fulfilled the criteria presented in Miller et al. (2010), Sahlée et al. (2011), Blomquist et al. (2014) and Landwehr et al. (2014). The CO_2 fluxes measured on R/V Aranda might be underestimated because the flow rate was too small, 10 slpm. No systematic differences were observed when the LI-7200 CO2 fluxes were compared to the LI-7500 on the ship mast and also to the measurements made at Harmaja.

page 13, line 395: "we calculated the expected statistical variability using the Co-spectrum". What does this mean - haven't heard of this technique before?

The statistical variability of the covariance (which equals the integral of the Co-spectrum) depends on the shape of the Co-spectrum. A white Co-spectrum implicates smaller statistical variability for the covariance than a peaked Co-spectrum. In our estimates we have taken the observed shape of the CO_2 Co-spectrum into account when calculating the estimate of the statistical variability of the covariance.

Comments on Figures

General comment on figures: many of them were difficult to read. Fonts were small, data points were small, color choices were difficult to discern. I had to have figures zoomed on a 27" monitor to interpret.

Fonts and curve thicknesses were enlarged in most of the figures.

Fig 1: suggest turning off satelite layer because large features that may be the bathymetry are easily construed as land. There is no scale shown on this map.

We improved the figure.

Fig 2. The yellow arrow hardly shows up - suggest making it more prominent.

The color of the arrow is now red and more prominent.

Fig. 3a. I do not have any height scales shown on the left for right axes. There appears to be a missing/broken streamline at the height of the second lowest probe height on the simulated mast on the right side. Why does the right side mast extend below the soil line to the same depth as the left side mast? Shouldn't it terminate just below the lowest streamline? What is the grey stippled shading near the bottom of the figure represent - land? What is the upside-down U-shaped object to the right of the right side mast?

We made a new figure to improve the quality of the figure and to make it clearer.

Fig 3b. The wind profiles are extrapolated from 6 m to the surface. How is this being done - they do not all appear to asymptote to (0,0)? Are you assuming some sort of roughness? My preference would be to stick to the data and not extrapolate, so profiles would extend 6-16 m.

Corrected.

Fig 3c What sort of fit is used to generate the dashed profiles? I would eliminate those curves and instead just connect the measured data points, similar to Fig 3a.

Corrected.

Fig 4. First 3 days 8/25-8/28 appear most ‘active’ in terms of the pollutant spikes or features in these time series in panels a,b,e. Then a precipitous drop at 8/28 around 1200 (precip?), after which fewer pollutant spikes.

No precipitation occurred. The reason for lower concentrations can be seen from Fig. 4c and Fig. 5; after noon 8/28 the western wind from Atlantic via Baltic Sea dominated carrying very clean air to Helsinki. For that reason it is important to show the trajectories in Fig. 5.

You could combine panels c and d using a little creativity - not clear why a separate panel is needed.

Panels c and d were merged.

Fig 5. How useful are these compared to the local wind patterns since the focus is on local emission sources which probably dominate the pollutant signals measured.

Please, see our response concerning Fig. 4.

Fig 8. panel a) no x-axis label provided. panel b) no x-axis label or units provided. The location of the shoreline could be added to these panels - it would be close to the right axis. The footprints are shown for neutral stability in both panels. Figure 9a shows that many (most?) periods were not neutrally stable (eg, abs(z/L)>0.1). Maybe one of these panels could be used to show the impact of non-neutral conditions on the footprint - both stable and unstable.

The labels, units and the shoreline were added to Fig. 8. The footprints in case of stable and unstable stratification are now shown in the supplement, Fig. S3.

Fig 11. panel a) what is the utility of the EC CO_2 flux measurements? Are these values reasonable? For example, what was pco2 in the seawater in the footprint? This could be used to assess whether the EC CO2 flux measurements are reasonable. Alternatively, they could assume a gas exchange coefficient (eg use Wanninkhof 2014) and compute what pCO2 was in the footprint. Are these pCO2 values reasonable?

The CO_2 fluxes were studied as a part of the ship emission study. However, Fig. 11a where the CO_2 and Ntot fluxes were presented as a function of wind direction was removed.
As stated in the text the results of the fluxes obtained at Harmaja during 2011 and 2012 were similar in magnitude than by Honkanen et al. (2018). Unfortunately, we did not have measurements of the content of CO_2 in the seawater in 2011. This was one of the reason to repeat the measurements in 2012 together with R/V Aranda where those measurements exist. It would be interesting to study how well the bulk models for the CO_2 transfer based on gradient data from open ocean describe the conditions in the coastal areas and archipelago. The transfer is controlled by turbulence from different sources: the formulas relying on wind speed as a proxy for the turbulence may not give the correct answers since the conditions in coastal areas differ from those in the open ocean. This would/will be an interesting study of its own, though.

Panel b shows that most EC CO2 fluxes are positive, so that surface water concentrations should be higher than air side. But there are several points (maybe 10%) where EC CO2 flux is negative. Did surface pCO2 concentrations decrease at these times, or were fluxes countergradient? (Ok - it looks like they are suggesting the noisier points are associated with ship-induced spikes in CO2 flux).

Direct measurements of the content of CO_2 in seawater were not available at the time of the measurement campaign.

Fig 12 panel a) comparison between EC CO2 flux from Harmaja and Aranda could be shown better. Add lines to the timeseries similar to Fig 12c. And add a scatter plot with Aranda on one axis and Harmaja on the other axis

Fig. 12a was corrected as suggested by the Editor.
The scatter plot was added, now Fig. 12b. The following text was added to:
page 15, lines 465-467: The scatter plot of the CO_2 fluxes at Harmaja and on R/V Aranda is presented in Fig. 12b. The large scatter is most probably due to the different locations: the measurements of pCO_2 (Fig. 12c) suggest that the pCO_2 was not spatially homogeneous. The estimated statistical variability is large and also contributes to the scatter.

panel b) Water pCO2 values of 900 ppm are large! That should provide a good flux signal. An interesting comparison would be to compute pCO2 using the EC CO2 flux measurements from Harmaja and a gas exchange coefficient (as suggested in comment on Fig 11), and to plot that with measured Aranda pCO2 concentration.

See the answer for the comments concerning Fig. 11a.

Follow-up points on your responses to referee #1 comments:
- Given their question about storage fluxes (and your response), should the fact that storage fluxes were not considered be added to the manuscript somewhere?

The following text was added to page 13, lines 387-388:
The storage fluxes were not considered at this campaign. The site was at the sea where most of the time the turbulent mixing was the driving force for gas and particle dispersion.

- Is it valuable to include Fig 1 from your responses to referee #1 in the supplement in case other readers have the same question?

The figure was added to the supplement, now Fig. S3, and the following text was added to page 14, lines 434--437.
We also made the comparison of GR and EC methods with sensible heat (Fig. S3). Clear correlation between the methods can be observed if the calculation of the sensible heat by GR method was conducted from the sea surface up the measurement height 11 m and to 15 m. However, the temperature difference between the measurement heights (11 and 15 m) were mostly too small to be detected and no flux could be calculated.

References:

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